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Exploring Novel Spintronic Responses from Advanced Functional Organic Materials

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Final Report

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14. ABSTRACT
The international collaborative project explored spin-dependent behaviors in excited states and charge transport based on unique conjugated organic materials. The research efforts have made major breakthroughs on molecular metamaterials by using spin radicals and on thermoelectric effects by using polymer/metal interface-controllable thermal and electric conductions. The project explored a new strategy by using interfacial polarization to address the challenging issue: separate controlling on electric and thermal conductions. This new strategy can lead to a significant enhancement on Seebeck effect.

15. SUBJECT TERMS
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Final Report

March 29, 2012 – March 28, 2015

Project title:

Exploring Novel Spintronic Responses from Advanced Functional Organic Materials

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Agreement Number:

AOARD-124007

Objectives:

The objective of this international collaborative project is to explore spin-dependent behaviors in excited states and charge transport based on unique conjugated organic materials.

Status of Effort:

The research efforts have made following major breakthroughs on molecular metamaterials by using spin radicals and on thermoelectric effects by using polymer/metal interface-controllable thermal and electric conductions.

1. Developed new strategy to couple semiconducting π electrons and magnetic d electrons for the development of molecular metamaterials [*Advanced Electronic Materials* DOI: 10.1002/aelm.201500058]

   The project has developed a new strategy for coupling semiconducting π electrons and magnetic d electrons by combining optically-generated intermolecular excited states with surface-modified magnetic nanoparticles. This new strategy can lead to optically-controllable composite metamaterials.

2. Explored new mechanism to utilize intermolecular excited states for realizing electric-magnetic coupling towards developing molecular metamaterials [*Adv. Mater* **26**, 3956-3961, 2014]

   The project has developed a new mechanism for realizing electric-magnetic coupling by using optically-generated intermolecular excited states in organic semiconducting materials. This new mechanism can generate optically-controllable molecular metamaterials.

3. Developed new method to use spin radicals for realizing electric-magnetic coupling towards radicals-based metamaterials [*Adv. Mater*. **23**, 2216-2220, 2011]

   The project has introduced spin radicals into organic semiconducting materials for the development of radicals-based metamaterials. This new method can develop spin-tunable electric-magnetic coupling for development of radicals-based metamaterials.

4. Discovered a novel mechanism to generate magneto-optic properties by establishing spin-exchange interaction in electron-hole pairs in ferroelectrically semiconducting materials [*Advanced Materials* **27**, 2899-2906, 2015]

   The project has discovered a novel mechanism to generate magneto-optic properties by establishing spin-exchange interaction in ferroelectrically semiconducting perovskites. This new mechanism leads to a breakthrough to create magneto-optic functions in all functional materials.

5. Developing new strategy to separately control electrical and thermal conductivities by using interfacial polarization [*Adv. Mater.* 23, 4120-4124, 2011]

The project explored a new strategy by using interfacial polarization to address the challenging issue: separate controlling on electric and thermal conductions. This new strategy can lead to a significant enhancement on Seebeck effect.

Accomplishments/New Findings:

I. Radicals-based molecular metamaterials

This research task has made the following five accomplishments including:

1. Discovery: new mechanism to realize electric-magnetic coupling for the development of molecular metamaterials

◆ **Enhanced electric-magnetic coupling from organic-magnetic nano-composite in excited state metamaterials** [*Advanced Electronic Materials* DOI: 10.1002/aelm.201500058]**.**

We found that a stronger electric-magnetic coupling between charge-transfer states and spin dipoles in electric-magnetic nano-composite can be realized when exciting the nano-composite into excited state. Fig. 1 shows an obvious enhanced amplitude and a line-shape change of magnetocapacitance in excited state from electric-magnetic nano-composite. Apparently, this striking mangetocapacitance change suggests that photoexcitation provide a new method to enlarge the electric-magnetic coupling between charge-transfer states and spin dipoles in the electric-magnetic nano-composite in excited state.

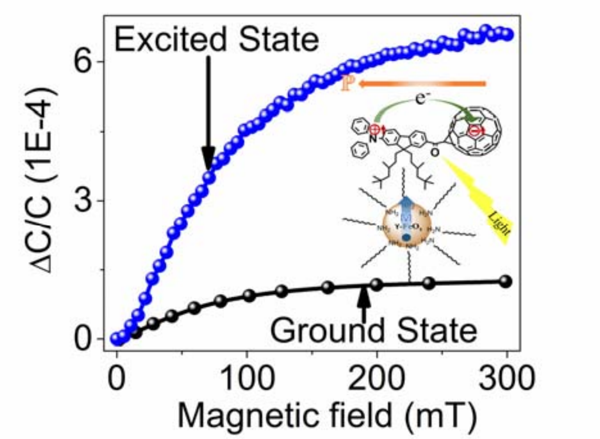


Fig. 1 Magnetocapacitance to show the larger electric-magnetic coupling in excited state.

◆ **Electric-magnetic coupling from radical pairs in organic molecules** [*Adv. Mater* **26**, 3956-3961, 2014].

We have discovered a new mechanism to generate electric-magnetic coupling by using radical pairs in organic semiconducting donor:acceptor systems. We can see in Fig. 2 that a pure semiconducting donor:acceptor (BBOT:TPD) demonstrates a significant magneto-dielectric function under photoexcitation: a magnetic field can change capacitance in organic semiconducting materials under photoexcitation. This magneto-dielectric function indicates a significant electric-magnetic coupling. Therefore, radical pairs present a new mechanism to generate electric-magnetic coupling towards the development of molecular metamaterials.

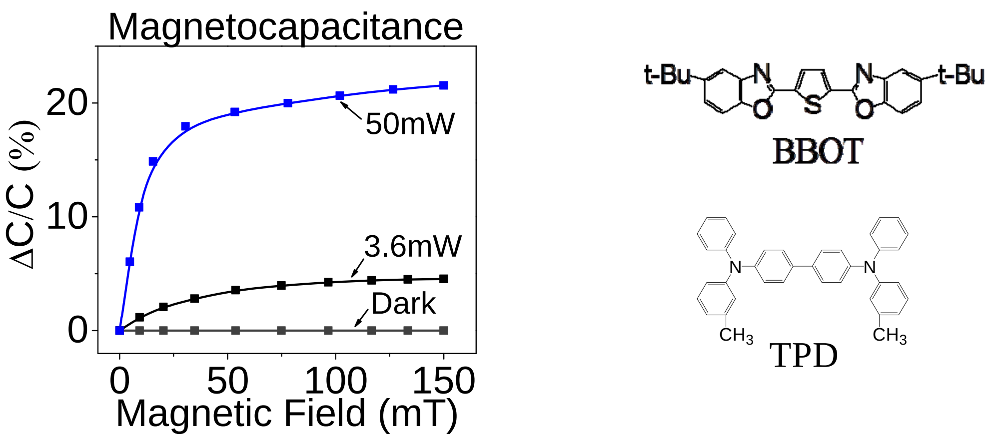
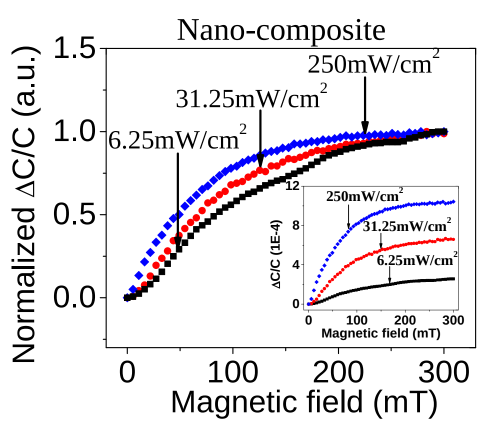


Fig. 2 Magnetocapacitance from optically-generated intermolecular excited states in semiconducting donor:acceptor (BBOT:TPD) composite.

2. New approach: Optically controlling magnetic properties by using charge-transfer states

◆ **Optically tunable magnetic properties through the interaction between intermolecular charge-transfer states and spin dipoles** [Submitted to *Advanced Electronic Materials*].

We have discovered that the magnetic properties of intermolecular charge-transfer states in electric-magnetic nano-composite can be adjusted by photoexcitation though varying the



Coulomb interaction between intermolecular charge-transfer states and magnetic spin dipoles. Fig. 3 depicts an interesting phenomenon of narrowing line-shape and increasing amplitude of magnetocapacitance with increasing photoexcitation intensities. As a consequence, this straightforward phenomenon clarify a unique way to tune the magnetic properties through changing the interaction between intermolecular charge-transfer states and magnetic spin dipoles with varied photoexcitation intensities.

Fig. 3 Magnetocapacitance in electric-magnetic nano-composite under different photoexcitation intensities.

◆ **Optically tunable magnetic properties through the interaction between intermolecular charge-transfer states** [*Adv. Mater* **26**, 3956-3961, 2014; *Physical Review B* 89, 155304 (2014)].

We have found that the spin-exchange energy in intermolecular charge-transfer states can be manipulated by the photoexcitation through changing the Coulomb interaction spin interaction among the intermolecular charge-transfer states. Fig. 4 indicates that the magnitude and line-shape of magnetocapacitance and magnetophotoluminescence can be changed by increasing the photoexcitation. Consequently, the line-shape narrowing of magnetocapacitance and magnetophotoluminescence illustrates a new way to tune the magnetic properties by photoexcitation through the interaction between intermolecular charge-transfer states, leading a new method to realize the molecular metamaterials.

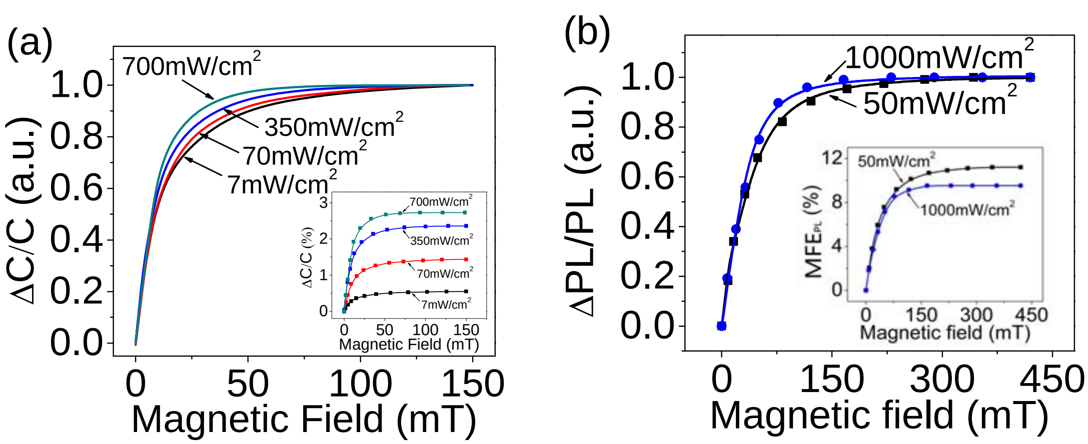


Fig. 4 (a) Magnetocapacitance in semiconducting donor:acceptor (BBOT:TPD) composite under different photoexcitation intensities. (b) Magnetophotoluminescence in semiconducting donor:acceptor (Pyrene:DMA) in DMF under different photoexcitation intensities.

3. Discovery: New approach to realize optically tunable plasmonics [to be published]

We have discovered a new mechanism to separately control the magnetized charge-transfer states and photo-induced charge-transfer states through the coupling interaction between them. Fig. 5 shows the coupling interaction between magnetized charge-transfer states and photo-induced charge transfer states becomes stronger with the decreasing distance. This new phenomenon on one hand provides a unique way to magnetically control the photo-induced charge-transfer states. On the other hand, it predicts a method to optically tune the magnetic plasmonic response. Consequently, this new finding affords a new approach to coupling the electric, optic and magnetic properties for developing new metamaterials.

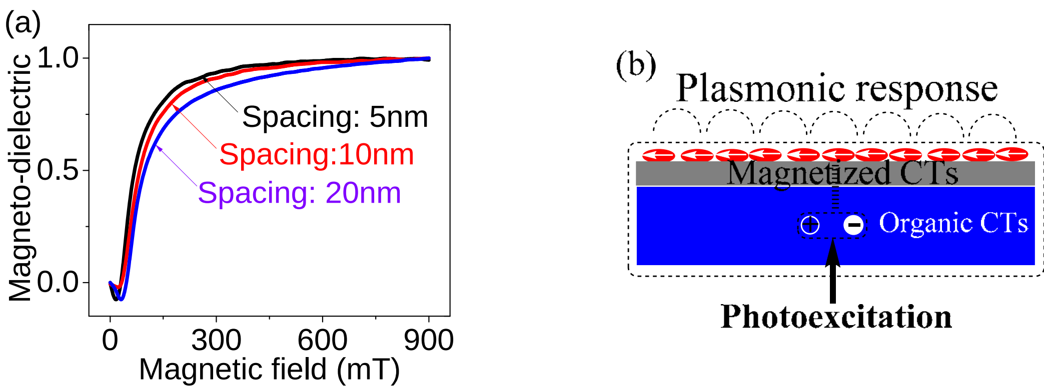


Fig. 5 (a) Magneto-dielectric property to show the coupling interaction between magnetized charge-transfer states and photo-induced charge transfer states. (b) Schematic for illustrating excitons-based plasmonics with optical tuning.

4. Discovery: Optically induced ferromagnetic properties from radical pairs [*Scientific reports* 5, 2015]

We have discovered a new phenomenon that electrogenerated chemiluminescence can produce a magnetic property after removing the applied magnetic field. Fig. 6 shows an abnormal magnetic field effect on electrogenerated chemiluminescence: the electrogenerated chemiluminescence can still present a change even without a magnetic field. This unexpected phenomenon suggests that the activated charge-transfer [Ru(bpy)33+…TPrA•] complexes may become magnetized in magnetic field and experience a long magnetic relaxation after removing magnetic field. Therefore, this new discovery presents a new way to manipulate the magnetic property for developing new metamaterials.

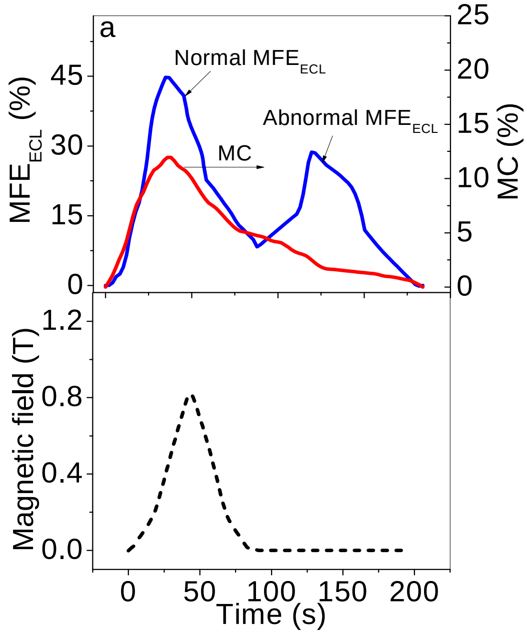


Fig. 6 (a) Magnetic field effect on electrogenerated chemiluminescence and magnetocurrent showing the magnetized charge-transfer states.

5. Discovery: Optically induced magneto-optic properties from spin-exchange interaction in ferroelectrically semiconducting materials [*Advanced Materials **27**, 2899-2906, 2015*

We have found that the photoluminescence and photocurrent can be changed by the external magnetic field through changing the spin-exchange interaction in ferroelectrically semiconducting perovskites. Fig. 7 indicates that the photoluminescence and photocurrent can be decreased or increased under the external magnetic field. Consequently, the change of photocurrent and photoluminescence illustrates a new way to tune the optical and electrical properties through tuning the spin-exchange interaction, leading to a breakthrough to create magneto-optic functions in all functional materials.

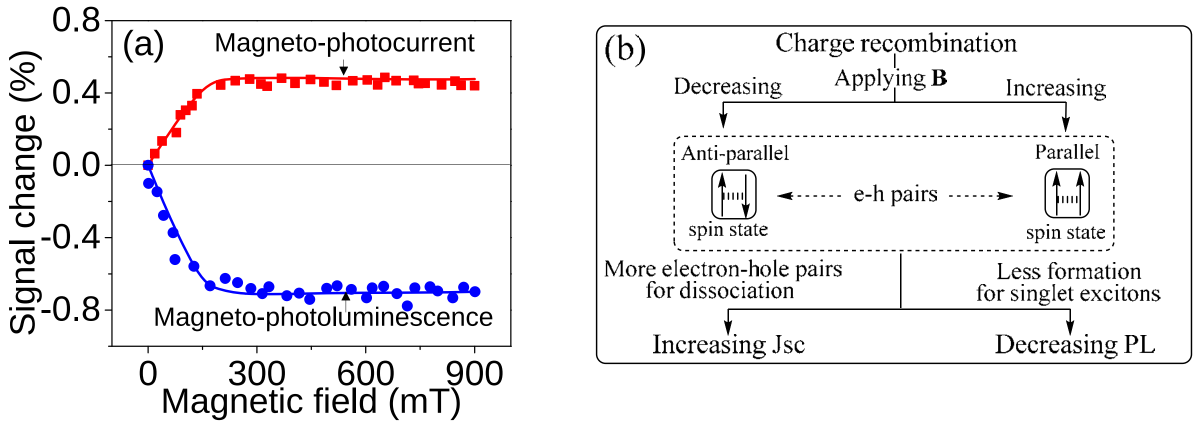


Fig. 7 (a) Magnetic field effect on photocurrent and photoluminescence. (b)Diagram to show the mechanism for magneto-optical effects in perovskites.

II. Thin-film based polymer thermoelectric devices

This research task has made the following six accomplishments including:

1. **New driving force of temperature-dependent surface polarization to cooperatively enhance Seebeck effect and electrical conductivity in vertical multilayer organic thin-film devices** [*Phys. Chem. Chem. Phys.* 16, 22201-22206, 2014]

We have explored a new mechanism to develop Seebeck effects by using temperature-dependent surface polarization based on vertical multi-layer thin-film devices (Al/P3HT:PCBM/Al, & Al/MoO_3/P3HT:PCBM/Al). Here, the temperature-dependent surface polarization functions as an additional driving force, as compared with the traditional driving force from entropy difference, to diffuse the charge carriers under temperature difference towards the development of Seebeck effects. We have demonstrate simultaneously enhanced Seebeck coefficient and electrical conductivity by using dielectric interface through the temperature-dependent surface polarization to diffuse charge carriers in the Al/MoO_3/P3HT:PCBM/Al thin-film device (Fig. 9). This temperature-dependent surface polarization provides a new mechanism allowing a co-operative relationship between Seebeck coefficient and electrical conductivity.

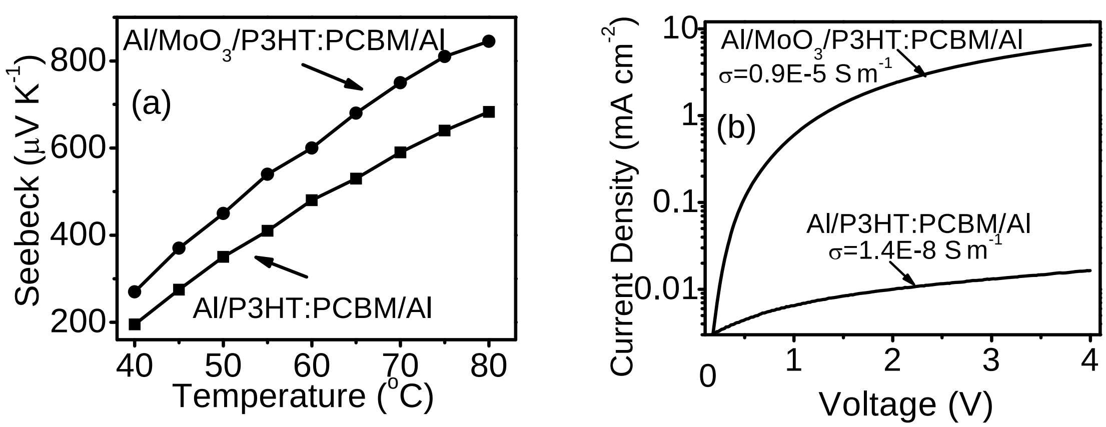


Fig. 9 (a) Seebeck coefficients in dark condition. (b) Electric conduction in dark condition.

2. **Dual functions of Seebeck and cooling effects in organic thin-film devices** [Submitted]

Here we have explored the possibilities of using temperature-dependent surface polarization as a new thermoelectric driving force to solve the conflicting requirement between electrical and thermal conductions in developing dual Seebeck and cooling effects based on the hybrid organic/inorganic Au/P(VDF-TrFE)/MoO_3/ITO thin-film device (Fig. 10). On one hand, the temperature-dependent surface polarization can lead to a temperature-dependent electrical field to drift charge carriers from high to low-temperature surface, generating a large Seebeck effect. On the other hand, the temperature-dependent surface polarization can absorb heat at the Au/organic interface through charge-phonon coupling by thermionic injection mechanisms when the charge carriers are injected upon applying an electrical bias, leading to a cooling effect. Essentially, the temperature-dependent surface polarization provides a mechanism to develop dual Seebeck and cooling effects through charge-phonon coupling based on thin-film electrode/organic/electrode devices.

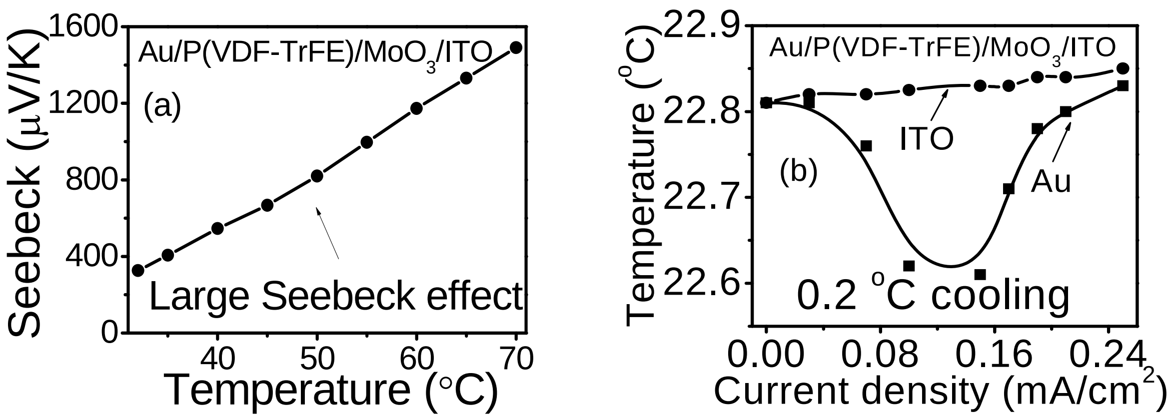


Fig. 10 (a) Seebeck coefficients. (b) Cooling effects with small injection currents.

3. Seebeck effects in n-type and p-type polymers driven simultaneously by surface polarization and entropy differences based on conductor/polymer/conductor thin-film devices [*ACS Nano*, 9, 5208-5213, 2015]

Here, we have reported a new approach of using intramolecular charge-transfer states to develop Seebeck effects driven by both surface polarization difference and entropy difference in n-type and p-type conjugated polymers based on vertical conductor/polymer/conductor thin-film devices. We have obtained larger Seebeck coefficients when the charge-transfer states are generated by a white light illumination (Fig. 11). Simultaneously, the electrical conductivities are increased from almost insulating state in dark condition to conducting state under photoexcitation in both n-type IIDT and p-type IIDDT based devices. Furthermore, we find that changing electrical conductivity can switch the Seebeck effects between polarization and entropy regimes when the charge-transfer states are generated upon applying photoexcitation. Therefore, using intramolecular charge-transfer states presents an approach to develop thermoelectric effects in organic materials-based vertical conductor/polymer/conductor thin-film devices.

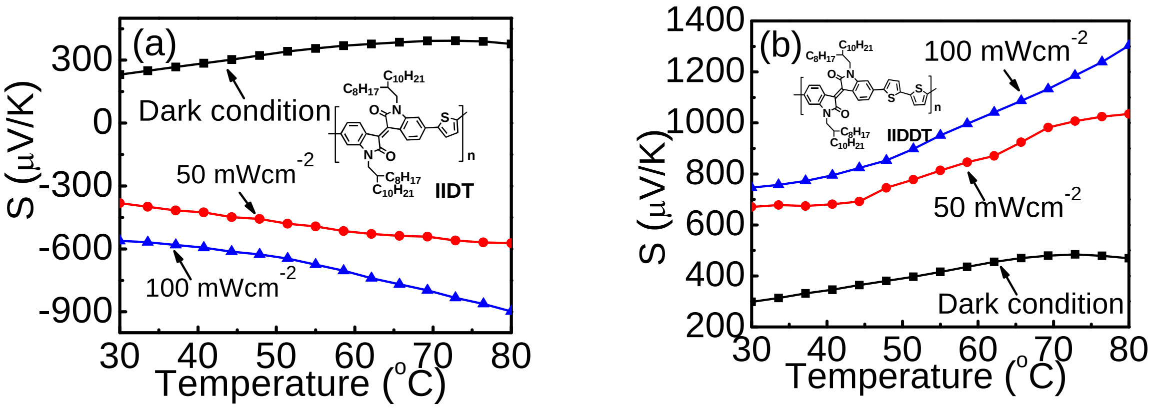


Fig. 11 Seebeck effects in n-type and p-type thin-film devices. (a) n-type ITO/IIDT/Au device. (b) p-type ITO/IIDDT/Au device.

4. New mechanism to generate magneto-Seebeck effect by applying Hall effect on organic thin-film devices [Submitted]

Hall effect can generate magneto-transport phenomenon in organic thin-film devices, and vertical organic thin-film devices can lead to large Seebeck effect. Therefore, combination of Hall effect and vertical organic thin-film devices can lead to a new mechanism to generate magneto-Seebeck effect. Here we have discovered giant magnetic field effects on Seebeck coefficients by applying Hall effect on vertical multi-layer ITO/PEDOT:PSS/Au thin-film devices (Fig. 12). This discovery demonstrates a magnetic approach to control the thermoelectric properties in organic thin-film devices.

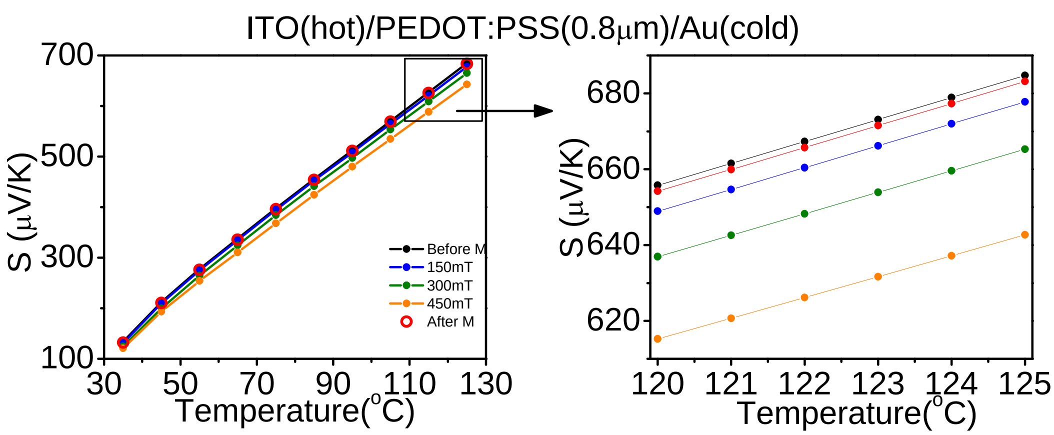


Fig. 12 Seebeck coefficient as a function of temperature under different magnetic field when magnetic field is perpendicular to temperature gradient.

Personnel Supported:

1. Ph.D student Mingxing Li with partial support
2. Ph.D student Qing Liu with partial support

Publication preparation from April 2012 to March 31 2015

1. Magneto-Optical Studies on Spin-Dependent Charge Recombination and Dissociation in Perovskite Solar Cells
   Yu-Che Hsiao, Ting Wu, Mingxing Li, and Bin Hu
   Adv. Mater. **27**. 2899, 2015
2. Enhanced π–d Electron Coupling in the Excited State by Combining Intramolecular Charge-Transfer States with Surface-Modified Magnetic Nanoparticles in Organic–Magnetic Nanocomposites
   Mingxing Li, Min Wang, Lei He, Yu-Che Hsiao, Qing Liu, Hengxing Xu, Ting Wu, Liang Yan, Loon-Seng Tan, Augustine Urbas, Long Y. Chiang and Bin Hu
   Adv. Electron. Mater. DOI: 10.1002/aelm.201500058, 2015
3. Abnormal Magnetic Field Effects on Electrogenerated Chemiluminescence
   Haiping Pan, Yan Shen, Hongfeng Wang, Lei He, Bin Hu
   Scientific Report. **5**, 9105, 2015
4. Changing the sign of exchange interaction in radical pairs to tune magnetic field effect on electrogenerated chemiluminescence
   Haiping Pan, Yan Shen, Lin Luan, Kai Lu, Jiashun Duan, and Bin Hu
   J. Phys. Chem. C. **119**, 8089, 2015
5. Dynamic Coupling between Electrode Interface and Donor/Acceptor Interface via Charge Dissociation in Organic Solar Cells at Device-Operating Condition
   Ting Wu, Yu-Che Hsiao, Mingxing Li, Nam-Goo Kang, Jimmy W. Mays, Bin Hu*
   J. Phys. Chem. C. **119**, 2727, 2015
6. Seebeck Effects in N-Type and P-Type Polymers Driven Simultaneously by Surface Polarization and Entropy Differences Based on Conductor/Polymer/Conductor Thin-Film Devices
   Dehua Hu, Qing Liu, Jeremy Tisdale, Ting Lei, Jian Pei, Hsin Wang, Augustine Urbas, and Bin Hu
   ACS Nano. **9**, 5208, 2015
7. Effects of a ferroelectric interface on thermionic injection-induced cooling in single-heterojunction devices based on thin-film electrode/medium/electrode design
   Qing Liu, Ting Wu, Yu-Che Hsiao, Mingxing Li, Dehua Hu, Hongfeng Wang, Hengxing Xu, Jeremy Tisdale and Bin Hu
   J. Mater. Chem. A. **3**, 14431, 2015

8. Optically tunable spin-exchange energy at donor:acceptor interfaces in organic solar cells
Mingxing Li, Hongfeng Wang, Lei He, Huidong Zang, Hengxing Xu and Bin Hu
Appl. Phys. Lett. **105**, 023302, 2014
9. Optically-Tunable Magneto-Capacitance Phenomenon in Organic Semiconducting Materials Developed by Electrical Polarization of Intermolecular Charge-Transfer States
Lei He, Mingxing Li, Augustine Urbas, and Bin Hu
Adv. Mater. **26**, 3956, 2014
10. Changing Line-Shape in Magnetic Field Effects through Interactions between Excited States in Organic Semiconducting Materials
Lei He, Mingxing Li, Augustine Urbas, and Bin Hu
Phys. Rev. B. **89**, 155304, 2014
11. Optically tunable spin-exchange energy at donor:acceptor interfaces in organic solar cells
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12. Inter-triplet spin-spin interaction effects on inter-conversion between different spin states in intermediate triplet-triplet pairs towards singlet fission
Xianfeng Qiao, Lin Luan, Yuchun Liu, Zhigang Yu, and Bin Hu
Org. Electron. **15**, 2168, 2014
13. Magneto-Dielectric Effects Induced by Optically-Generated Intermolecular Charge-Transfer States in Organic Semiconducting Materials
Huidong Zang, Liang Yan, Mingxing Li, Lei He, Zheng Gai, Ilia Ivanov, Min Wang, Long Chiang, Augustine Urbas, and Bin Hu
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Huidong Zang, Jianguo Wang, Mingxing Li, Lei He, Zitong Liu, Deqing Zhang, and Bin Hu
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15. Enhancing Seebeck Effects by Using Excited States in Organic Semiconducting Polymer MEH-PPV Based on Multi-layer Electrode/Polymer/Electrode Thin-Film Structure
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J. Phys. Chem. C. **117**, 10264, 2013
16. Triplet-Charge Annihilation versus Triplet-Triplet Annihilation in Organic Semiconductors
Ming Shao, Liang Yan, Li Mingxing, Ivanov Ilia, and Bin Hu
J. Mater. Chem. C. **1**, 1330, 2013
17. High Seebeck Effects from Conducting Polymer: Poly(3,4-ethylenedioxythiophene): Poly(styrenesulfonate) Based Thin-Film Device with Hybrid Metal/Polymer/Metal Architecture
Michael Stanford, Hsin Wang, Ilia Ivanov, and Bin Hu
Appl. Phys. Lett. **101**, 173304, 2012

18. Magnetocurrent of Charge-Polarizable C_{60}-Diphenylaminofluorene Monoadduct-Derived Magnetic Nanocomposites
Liang Yan, Min Wang, N.P. Raju, Arthur Epstein, Loon-Seng Tan, Augustine Urbas, Long Y. Chiang, Bin Hu
J. Am. Chem. Soc. **134**, 3549-3554, 2012
19. Changing Inter-molecular Spin-Orbital Coupling for Generating Magnetic Field Effects in Phosphorescent Organic Semiconductors
Liang Yan, Ming Shao, Carlos F. O. Graeff, Ivo Hummelgen, Dongge Ma, Bin Hu
Appl. Phys. Lett. **100**, 013301, 2012

Interactions and Transitions:

a. Presentations at Conferences

Invited Presentation from July 01 2011 to December 31 2014

(1) **Magneto-Optical Studies on Organic and Perovskite Solar Cells**
Bin Hu
Asian Conference on Organic Electronics, National Cheng Kung University, Tainan, Taiwan, November 12-15, 2014

(2) **Magneto-Dielectric Effects Generated by Charge-Transfer States in Organic Semiconductors**
Bin Hu
5th Topical Meeting on Spins in Organic Semiconductors, Himeji, Japan, October 14-17, 2014

(3) **New Magnetic Field Effects in Organic Semiconductors**
Bin Hu
2014 International Symposium on Materials for Enabling Nanodevices, National Cheng Kung University, Tainan, Taiwan, September 03-06, 2014

(4) **Magneto-optic properties in organic materials**
Bin Hu
AOARD conference on magnetic nanomaterials, University of Maryland, June 16-17, 2014

(5) **Organic spintronics, organic solar cells, and organic thermoelectrics**
Bin Hu
E-MRS, Lille, France, May 26-30, 2014

(6) **Magneto-optic properties in organic materials**
Bin Hu
US-Taiwan Air Force Conference, Hualien, Taiwan, May 13-15, 2014

(7) **Magneto-optic studies of photovoltaic processes at D:A interface and electrode interface in organic solar cells**
Bin Hu
Indo-US Joint Workshop on Organic Solar Cells, Kanpur, India, March 20-22, 2014

(8) **Interface enhanced photovoltaic and Seebeck effects in organic solar cells and thermoelectric devices**

Bin Hu
ACS Annual Meeting, Dallas, TX, March 16, 2014

(9) Multiferroic Effects from Intermolecular Excited States in Organic Semiconductors
Bin Hu
Brazil-MRS meeting, Campos do Jordao, September 30 – October 04, 2013

(10) Magneto-Optic, Magneto-Electric, and Magneto-Thermoelectric Effects in Organic Semiconductors
Bin Hu
BES Program Review for the CNMS at Oak Ridge National Laboratory, September 24-26, 2013

(11) Organic Thin-Film Thermoelectric Devices
Bin Hu
Flexible Thermoelectric Workshop organized by AFOSR, Arlington, VA, July 09-10, 2013

(12) Effects of Intermolecular and Dielectric-layer Interfaces on Internal Photovoltaic Processes in Organic Solar Cells
Bin Hu
Indo-US Joint Workshop on Organic Solar Cells, National Reviewable Energy Laboratory, Golden, Co, June 24-25, 2013

(13) Magneto-optical Studies on Internal Photovoltaic Processes in Organic Solar Cells
Bin Hu
2013 TechConnect World, National Innovation Summit and National SBIR Conference, Gaylord Hotel, National Harbor, Maryland, May 13-16, 2013

(14) Magneto-Dielectric Functions Developed by Intermolecular Excited States
Bin Hu
MRS Meetings, San Francisco, CA, April 01-05, 2013

(15) Departmental Seminar: Organic Spintronics
Bin Hu
National Taiwan University, Taipei, Taiwan, December 11, 2012

(16) Workshop on Organic Spintronics
Bin Hu
Intermolecular Excited States-Based Organic Spintronics
National Cheng Kung University, Tainan, Taiwan, December 06-07, 2012

(17) Effects of Intermolecular and Dielectric-layer Interfaces on Internal Photovoltaic Processes in Organic Solar Cells
Bin Hu
International Symposium on Organic and Dye-Sensitized Solar Cells 2012 (IS-OPVDSC 2012), Taipei, Taiwan, November 24-29, 2012

(18) Electric-Magnetic Coupling in Organic Sintronics
Bin Hu
9th National Conference on Organic Solids Electronics, Yangzhou, China, November 10-12, 2012

(19) Magneto-optical studies on internal photovoltaic processes in organic solar cells
Bin Hu

Workshop on key scientific and technological issues for development of next-generation organic solar cells
Arlington, VA, September 20 – 21, 2012

(20) **Multi-Ferroic Functions Developed by Inter-molecular Excited States**
Bin Hu
4th Topical Meeting on Spintronics in Organic Semiconductors, London, UK, September 10 – 14, 2012

(21) **Excited States-Based Organic Spintronics**
Bin Hu
International Workshop on Novel Nano-Magnetic and Multifunctional Materials 2012
Seoul, Korea, June 11-14, 2012

(22) **Magneto-Optical Studies of Internal Photovoltaic Processes in Organic Solar Cells**
Bin Hu
Departmental seminar at Department of Materials Science and Engineering, University of Florida, Gainesville, FL, April 04, 2012

(23) **Organic Molecular Metamaterials**
Bin Hu
Organic Metamaterials Workshop, Army Research Laboratory, March 02, 2012

(24) **Characterization and Understanding on Internal Photovoltaic Processes in Organic Solar Cells**
Bin Hu
International Photonics Conference – 2011, Tainan, Taiwan, December 07-08, 2011

(25) **Organic Spintronics: Magnetic Field Effects**
Bin Hu
International Symposium on Organic Dye Sensitized Solar Cells, Tainan, Taiwan, December 08-10, 2011

(26) **The Role of Inter-molecular Electron-Hole Pairs in Magnetic Field Effects in Organic Materials**,
Bin Hu
61st Annual Meeting of Japan Coordination Chemistry Society, Okayama, Japan, September 17-19, 2011

(27) **Characterization and Understanding on Charge Dissociation, Transport, Collection in Organic Solar Cells**
Bin Hu
Workshop on Sustainable Energy Future: Focus on Organic Photovoltaics, Oak Ridge National Laboratory, September 21 – 23, 2011

(28) **Electromagnetic and Thermoelectric Responses from Inter-molecular Excited States: Radicals Pairs**
Bin Hu
8th US-Taiwan NanoScience Workshop, Seattle, Washington, April 05-06, 2011

b. *Consultative and advisory functions*

None.

c. *Transitions*

None.

New Discoveries

Four experimental discoveries have been made. They include:

(1) New strategy to couple semiconducting π electrons and magnetic d electrons for development of molecular metamaterials
(2) New mechanism to use intermolecular excited states for realizing electric-magnetic coupling towards development of molecular metamaterials
(3) New method to use spin radicals for realizing electric-magnetic coupling towards radicals-based metamaterials
(4) New mechanism to develop significant magneto-optic properties by combining Hall effect with spin nradicals
(5) New strategy to separately control electrical and thermal conductivities by using interfacial polarization

Honors/Awards:

Research Achievement Award – April 2014
College of Engineering
University of Tennessee

Research Fellow Award – April 2012
College of Engineering
University of Tennessee

Publication list – Bin Hu

AOARD project: Exploring Novel Spintronic Responses from Advanced Functional Organic Materials

1. Magneto-Optical Studies on Spin-Dependent Charge Recombination and Dissociation in Perovskite Solar Cells
   Yu-Che Hsiao, Ting Wu, Mingxing Li, and Bin Hu
   Adv. Mater. **27**. 2899, 2015
2. Enhanced π–d Electron Coupling in the Excited State by Combining Intramolecular Charge-Transfer States with Surface-Modified Magnetic Nanoparticles in Organic–Magnetic Nanocomposites
   Mingxing Li, Min Wang, Lei He, Yu-Che Hsiao, Qing Liu, Hengxing Xu, Ting Wu, Liang Yan, Loon-Seng Tan, Augustine Urbas, Long Y. Chiang and Bin Hu
   Adv. Electron. Mater. DOI: 10.1002/aelm.201500058, 2015
3. Abnormal Magnetic Field Effects on Electrogenerated Chemiluminescence
   Haiping Pan, Yan Shen, Hongfeng Wang, Lei He, Bin Hu
   Scientific Report. **5**, 9105, 2015
4. Changing the sign of exchange interaction in radical pairs to tune magnetic field effect on electrogenerated chemiluminescence
   Haiping Pan, Yan Shen, Lin Luan, Kai Lu, Jiashun Duan, and Bin Hu
   J. Phys. Chem. C. **119**, 8089, 2015
5. Dynamic Coupling between Electrode Interface and Donor/Acceptor Interface via Charge Dissociation in Organic Solar Cells at Device-Operating Condition
   Ting Wu, Yu-Che Hsiao, Mingxing Li, Nam-Goo Kang, Jimmy W. Mays, Bin Hu*
   J. Phys. Chem. C. **119**, 2727, 2015
6. Seebeck Effects in N-Type and P-Type Polymers Driven Simultaneously by Surface Polarization and Entropy Differences Based on Conductor/Polymer/Conductor Thin-Film Devices
   Dehua Hu, Qing Liu, Jeremy Tisdale, Ting Lei, Jian Pei, Hsin Wang, Augustine Urbas, and Bin Hu
   ACS Nano. **9**, 5208, 2015
7. Effects of a ferroelectric interface on thermionic injection-induced cooling in single-heterojunction devices based on thin-film electrode/medium/electrode design
   Qing Liu, Ting Wu, Yu-Che Hsiao, Mingxing Li, Dehua Hu, Hongfeng Wang, Hengxing Xu, Jeremy Tisdale and Bin Hu
   J. Mater. Chem. A. **3**, 14431, 2015

8. Optically tunable spin-exchange energy at donor:acceptor interfaces in organic solar cells
   Mingxing Li, Hongfeng Wang, Lei He, Huidong Zang, Hengxing Xu and Bin Hu
   Appl. Phys. Lett. **105**, 023302, 2014

9. Optically-Tunable Magneto-Capacitance Phenomenon in Organic Semiconducting Materials Developed by Electrical Polarization of Intermolecular Charge-Transfer States
Lei He, Mingxing Li, Augustine Urbas, and Bin Hu
Adv. Mater. **26**, 3956, 2014
10. Changing Line-Shape in Magnetic Field Effects through Interactions between Excited States in Organic Semiconducting Materials
Lei He, Mingxing Li, Augustine Urbas, and Bin Hu
Phys. Rev. B. **89**, 155304, 2014
11. Optically tunable spin-exchange energy at donor:acceptor interfaces in organic solar cells
Mingxing Li, Hongfeng Wang, Lei He, Huidong Zang, Hengxing Xu, and Bin Hu
Appl. Phys. Lett. **105**, 023302, 2014
12. Inter-triplet spin-spin interaction effects on inter-conversion between different spin states in intermediate triplet-triplet pairs towards singlet fission
Xianfeng Qiao, Lin Luan, Yuchun Liu, Zhigang Yu, and Bin Hu
Org. Electron. **15**, 2168, 2014
13. Magneto-Dielectric Effects Induced by Optically-Generated Intermolecular Charge-Transfer States in Organic Semiconducting Materials
Huidong Zang, Liang Yan, Mingxing Li, Lei He, Zheng Gai, Ilia Ivanov, Min Wang, Long Chiang, Augustine Urbas, and Bin Hu
Scientific Report **3**, 2812, 2013
14. Spin Radicals Enhanced Magnetocapacitance Effect in Intermolecular Excited States
Huidong Zang, Jianguo Wang, Mingxing Li, Lei He, Zitong Liu, Deqing Zhang, and Bin Hu
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15. Enhancing Seebeck Effects by Using Excited States in Organic Semiconducting Polymer MEH-PPV Based on Multi-layer Electrode/Polymer/Electrode Thin-Film Structure
Ling Xu, Yuchun Liu, Matthew P.Garrett, Bingbing Chen, and Bin Hu
J. Phys. Chem. C. **117**, 10264, 2013
16. Triplet-Charge Annihilation versus Triplet-Triplet Annihilation in Organic Semiconductors
Ming Shao, Liang Yan, Li Mingxing, Ivanov Ilia, and Bin Hu
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17. High Seebeck Effects from Conducting Polymer: Poly(3,4-ethylenedioxythiophene): Poly(styrenesulfonate) Based Thin-Film Device with Hybrid Metal/Polymer/Metal Architecture
Michael Stanford, Hsin Wang, Ilia Ivanov, and Bin Hu
Appl. Phys. Lett. **101**, 173304, 2012

18. Magnetocurrent of Charge-Polarizable C_{60}-Diphenylaminofluorene Monoadduct-Derived Magnetic Nanocomposites
Liang Yan, Min Wang, N.P. Raju, Arthur Epstein, Loon-Seng Tan, Augustine Urbas, Long Y. Chiang, Bin Hu
J. Am. Chem. Soc. **134**, 3549-3554, 2012

19. Changing Inter-molecular Spin-Orbital Coupling for Generating Magnetic Field Effects in Phosphorescent Organic Semiconductors
Liang Yan, Ming Shao, Carlos F. O. Graeff, Ivo Hummelgen, Dongge Ma, Bin Hu
Appl. Phys. Lett. **100**, 013301, 2012

1.

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Grant/Contract Number
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FA2386-12-1-4007

Principal Investigator Name
The full name of the principal investigator on the grant or contract.

Bin Hu

Program Manager
The AFOSR Program Manager currently assigned to the award

Fumiko Kano

Reporting Period Start Date

03/29/2012

Reporting Period End Date

03/28/2015

Abstract

The international collaborative project explored spin-dependent behaviors in excited states and charge transport based on unique conjugated organic materials. The research efforts have made major breakthroughs on molecular metamaterials by using spin radicals and on thermoelectric effects by using polymer/metal interface-controllable thermal and electric conductions. The project explored a new strategy by using interfacial polarization to address the challenging issue: separate controlling on electric and thermal conductions. This new strategy can lead to a significant enhancement on Seebeck effect.

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AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
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Total			

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Appendix Documents

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